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ENERGY TRANSFER IN SOLID LASER MATERIALS

Technical Summary Report No. 1

1 May - 31 July 1964

Contract Nonr 225(78)

ARPA Order No. 306-64

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INTRODUCTION

This report is the First Technical Summary Report under Contract Nonr 225(78), which began on 1 May 1964, and it reports the period of 1 May through 31 July 1964. At the present time there are 3 projects active under this contract:

1. Ultraviolet excitation of ruby
 - A. Nature of the excited states in ruby
 - B. Energy transfer between levels and between ions in ruby
2. Energy transfer between unlike ions in lanthanum fluoride

The Responsible Investigator for this contract is A. L. Schawlow.

OBJECTIVE

The purpose of this investigation is to study methods of exciting solid materials for visible, and possibly ultraviolet, maser action, and of energy transfer processes between energy levels and between ions.

I. ULTRAVIOLET EXCITATION OF RUBY

It is well known that, at low levels of excitation, the characteristic R-line fluorescence can be excited by ultraviolet radiation. The quantum efficiency for this process has been measured and found to be high.¹ Thus, it might appear that ultraviolet light, as provided by double-pulsed flash lamps,² would be suitable for pumping ruby lasers. If so, this would be very useful, because as lamp currents are increased to provide greater brightness, the increase is most marked in the ultraviolet region.

A. NATURE OF THE EXCITED STATES IN RUBY

(G. K. Klauminzer, H. W. Moos, P. L. Scott)

One possible difficulty in using ultraviolet light for laser pumping is that, as Maiman,³ F. Gires and G. Mayer have shown, when ruby is excited vigorously enough to produce appreciable population in the 2E state, strong new ultraviolet absorption bands appear. The obvious explanation is that the new absorptions correspond to upward transitions from the metastable 2E state to higher states of the Cr^{3+} ion. Thus, the new absorptions should all occur at wavenumbers $14,400\text{ cm}^{-1}$ less than known absorptions from the ground state to the same upper levels. However, the absorption bands based on the 2E level, as found by Gires and Mayer, do

¹E. E. Bukke and Z. L. Morgenstern, Optics and Spectroscopy 14, 362 (1963).

²J. L. Emmett and R. W. Hellworth, Bull. Am. Phys. Soc., Series II 7, 615 (1962); J. L. Emmett and A. L. Schawlow, Appl. Phys. Lett. 2, 204 (1963).

³T. H. Maiman, Phys. Rev. Lett. 4, 564 (1960); F. Gires and G. Mayer, Ann. Radioelect. 18, 112 (1963).

not look much like the absorption from the ground 4A_2 level shifted $14,400\text{ cm}^{-1}$ to the red. Indeed, some strong bands appear to have no counterpart in the ordinary absorption spectrum. Such considerations have led Schultz⁴ to suggest a radically different explanation of the bands. He considers the chromium ions in the ground state and in the excited state as two different species of ion, each modifying its surroundings in a characteristic way.

Despite the different appearance of the absorption spectra from the 4A_2 and 2E levels, the original explanation of Gires and Mayer may be correct. After all, the selection rules from these states could be, indeed must be, different. In particular, transitions from 4A_2 to all excited doublet states are spin forbidden and therefore weak. Transitions from 2E to excited doublet states, on the other hand, will be relatively strong. Thus, the transitions from 4A_2 to high doublet states which are apparently missing might be found if a sufficiently long absorption path is used, and indeed some of them are found. However, weak broad bands are easily obscured by overlapping stronger bands.

A good test of the alternative theories could come from the observation of sharp-line absorption from the 2E level to higher sharp levels, such as the three components of 2T_2 . The wavenumbers of these transitions could be measured with some accuracy. In this way, we could determine whether the transitions are exactly at the position given by the difference of distances from the ground state.

This experiment is now in progress. A ruby crystal is excited by a flash lamp, and is traversed by infrared radiation near 5500 cm^{-1} , corre-

⁴G. V. Schultz, Phys. Letters 8, 93 (1964).

sponding to the transition ${}^2E \rightarrow {}^2T_2$. This radiation passes through a Jarrel-Ash one meter high-resolution spectrometer which is equipped with a reasonably fast infrared detector. There is a filter in the spectrometer to block stray visible and near-infrared light from the flashlamp.

When the crystal is pumped by the flashlamp to provide some population of Cr^{3+} ions in the 2E level, new sharp absorptions appear in the expected region. Positions and widths of these lines correspond closely to those expected for the transition ${}^2E \rightarrow {}^2T_2$. While this work is still in progress, it appears tentatively that the upward excitation from the 2E level can be understood for the most part in terms of the levels discovered by ground-state absorption. Schultz's explanation seems unnecessary, at least for the lower excited levels.

B. ENERGY TRANSFER BETWEEN LEVELS AND BETWEEN IONS IN RUBY

(J. L. Emmett, R. L. Greene, S. A. Johnson, W. M. Yen)

The existence of the new ultraviolet absorption bands in ruby, which appear when the metastable 2E state is populated, presents two apparent obstacles to the use of ultraviolet light for pumping ruby. First, the strong absorption makes it difficult for some wavelengths of the ultraviolet light to penetrate to the interior of the laser rod. Second, when ultraviolet is absorbed in these new bands, Cr^{3+} ions are excited upward out of the desired 2E state. Whether these difficulties are serious or not must be investigated. Thus, when a chromium ion is excited upward from the 2E state, it returns almost instantaneously. Moreover, it is quite possible that the ion excited in this way may use its excess energy to excite a neighboring ion from the ground state to the metastable state.

The strong absorption of the new bands will indeed prevent some, although not all, wavelengths of ultraviolet light from reaching the center of the crystal once the 2E state is appreciably populated. However, there may well be some nonradiative spatial diffusion of excitation inward from the surface. There is very little knowledge of spatial diffusion rates, and we are planning experiments to investigate them.

The most direct test of the usefulness of ultraviolet light will be to try to pump a ruby laser with only ultraviolet light. A materials testing setup using a double-pulsed flashlamp to provide intense ultraviolet light, and with provision for filtering, is under construction. When completed, it will be available for this program, and will be used for tests of ultraviolet excitation.

One situation where energy transfer surely occurs is between isolated chromium ions and ion pairs in ruby. This transfer must be invoked to account for the anomalously large fluorescence intensity from the ion pairs.⁵ It also makes possible laser action at 7009 Å and 7041 Å from ion pairs in dark ruby.

However, it has not been shown whether or not the energy transfer to the paired ions occurs from isolated chromium ions in the 2E state. It is possible that most of the transfer takes place from ions in the 4T_1 and 4T_2 states to which they are initially pumped. Such a process is suggested by the fact that the R-lines (isolated ions) and N-lines (paired ions) do not have exactly the same radiative lifetime. However, selective excitation experiments by Daly show that radiation in the neighborhood of the R-lines can excite the N-lines.

We plan to investigate the energy transfer between isolated ions in the 2E state and paired ions by suddenly exciting the isolated chromium ions with intense radiation at the R_1 wavelength. This will be done by a Q-spoiled ruby laser. This laser, using a rotating prism for Q-spoiling, is now operating. In the near future, it will be applied in an attempt to excite the N-line fluorescence in a piece of dark ruby. Then the kinetics of energy transfer from the 2E state of isolated ions to ion pairs will be studied.

⁵A. L. Schawlow, D. L. Wood and A. M. Clogston, Phys. Rev. Lett. 3, 544 (1959); G. E. Devlin and A. L. Schawlow, Phys. Rev. Lett. 6, 96 (1961).

II. ENERGY TRANSFER BETWEEN UNLIKE IONS IN LANTHANUM FLUORIDE (W. C. Scott, W. M. Yen)

Recently, widths of emission and absorption lines of praseodymium (Pr^{3+}) ions in LaFs have been investigated.⁶ It was found that in an isolated group of levels at low temperatures, the lowest showed only strain broadening. The spectral line joining such a level to the ground state shows the Gaussian shape characteristic of strain broadening. In a good crystal, such a line may be quite narrow, less than 1 cm^{-1} in width.

Levels spaced $10\text{-}500 \text{ cm}^{-1}$ above such a level are usually broader because these levels can relax to each other and to the lowest of the group. This relaxation can occur in times of the order of from 10^{-11} to 10^{-12} sec, leading to line widths of the order of $1\text{-}10 \text{ cm}^{-1}$. However, the lowest level of the group has no other level into which it can empty, and so is narrow.

This same sharp line has now been studied in crystals containing various amounts of neodymium (Nd^{3+}) in addition to Pr^{3+} . The neodymium ion has a level only $\sim 100 \text{ cm}^{-1}$ below the lowest Pr^{3+} level of the group. It is found that the sharp Pr^{3+} line broadens and changes from a Gaussian to a Lorentzian shape as the Nd^{3+} concentration is increased.

This change in linewidth and character indicates that the Nd^{3+} ion provides a lower level to which the Pr^{3+} ion can relax. In the relaxation process, the excess energy is delivered to the crystal lattice.

⁶W. M. Yen, W. C. Scott and A. L. Schawlow, Phys. Rev., to be published.

Previous theoretical studies of interionic energy transfer have assumed that the ions have the same energy levels, so that their transitions are resonant. Thus energy is transferred from one to the other without considering the lattice vibrations. We have considered the case where there is a difference energy to be delivered to the lattice vibrations. It is found that energy transfer rates of the order of those observed can be explained.